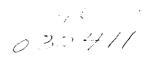
American Mineralogist, Volume 58, pages 721-726, 1973



# Al-Rich Pargasite

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#### Abstract

Two Al-rich amphiboles were found in a corundum- and spinel-bearing calcite marble together with minor phlogopite, margarite, anorthite, and sphene or rutile. Microprobe analyses give half their unit cell contents as:

Amphibole AA-5,  $Na_{0.54}K_{0.16}Ca_{1.99}Mg_{3.35}Fe_{0.01}Ti_{0.16}Al_{1.14}^{VI}Al_{2.26}^{VI}Si_{5.74}OH_{1.4}F_{0.40}Cl_{0.07}$ ; Amphibole HA-1/2,  $Na_{0.67}K_{0.17}Ca_{1.99}Mg_{3.62}Fe_{0.07}Ti_{0.27}Al_{1.12}^{VI}Al_{2.25}^{VI}Si_{5.75}OH_{1.05}F_{0.58}Cl_{0.02}$ . Amphibole AA-5 is the most aluminous yet found in nature (22.6 wt percent  $Al_2O_3$ ) and contains essentially no Fe (total iron as FeO = 0.05 wt percent).

Both amphiboles are too rich in  $Al^{1V}$  (viz, high  $Al_2O_3$  combined with low  $SiO_2$ ) to consider them as mixed crystals strictly intermediate between pure pargasite and pure tschermakite. Al-rich pargasite appears to be a suitable name for amphiboles of this composition, owing to the fact that more than 50 percent of their structural formulae can be recalculated in terms of a hypothetical end-member molecule Al-pargasite,  $(Na,K)Ca_2Mg_3Al^{VI}_2$   $Al^{IV}_3$   $Si_5O_{22}(OH,F)_2$ , which conforms to the limiting  $Al^{VI}$  ratio of Ca-amphiboles statistically evaluated by Leake (1965, 1971).

The water determination and X-ray powder data are given for one of the amphiboles but not for the other owing to insufficient material. Conditions for the crystallization of the Al-rich amphiboles are briefly discussed.

#### Introduction

The maximum Al-content possible in calciferous and subcalciferous amphiboles is of considerable interest for ascertaining crystallization environments. So far, no hornblende approaching the tschermakite (ferrotschermakite) end member, Ca2 (Mg, Fe2+)3 AlVI2AlIV2Si6O22(OH, F)2 as defined by Winchell (1945), has been detected in nature. Even amphiboles intermediate between tschermakite (ferro-tschermakite) and pargasite (hastingsite), NaCa2(Mg,  $Fe^{2+}$ <sub>4</sub> $Al^{VI}Al^{IV}$ <sub>2</sub> $Si_6O_{22}(OH, F)_2$ , are rare. From 1217 Ca-amphibole analyses compiled by Leake (1968), not more than 31 revealed Si < 6.25 (Aliv > 1.75) and Al<sup>vi</sup> > 1.00 per half-unit cell ( $\Sigma = >2.75$  Al), and only four of them gave Alvi > 1.30. Moreover, these four analyses were discredited by Leake (1968) who assumed that the extremely high AlVI contents were due either to analytical errors or to insufficient

mineral separation. The same holds true for four analyses from Leake's compilation with Alvi > 1.30, but  $Al^{IV}$  < 1.75 (Si > 6.25). In evaluating his compilation, Leake (1968, 1971) concluded that in calciferous amphiboles "the maximum possible  $Al^{vI}$  at Si = 6.00,  $Al^{IV} = 2.00$ , seems to be about 1.40." A necessary requisite for the formation of Alrich calcium amphiboles is a highly aluminous environment. In addition, AlVI substitution is favored by increasing pressure (Leake, 1965, 1971; Kostyuk and Sobolev, 1969). The reliably analyzed amphibole most closely approaching the tschermakite end-member is from a kyanite-garnet-biotite-hornblende gneiss from Frodalera (Lukmanier area, Switzerland) which indicates a high pressure, high-Al metamorphic environment. The analysis of this amphibole by Frey (1969), slightly modified by Leake (1971, Table 1), yields Si, 6.22; Aliv, 1.78; Alvi, 1.35; Na + K, 0.49; and Ca + Na + K, 2.15. Logically, it would seem promising to find high-AlVI-amphiboles in corundumhornblende rocks, provided the pressure of metamorphism was not too low. However, none of the

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amphiboles from recently analyzed corundum-bearing rock (Forestier and Lasnier, 1969; Leake, 1971) revealed extraordinarily high alumina contents. The most Al-rich amphibole within this group, from a corundum-zoisite-amphibole rock at Merkerstein (N. Arusha, Tanzania), is rather close to the theoretical pargasite composition: Si, 6.05; Al<sup>1V</sup>, 1.95; Al<sup>VI</sup>, 1.04; Na + K, 0.79, and Ca + Na + K, 2.60 (Leake, 1971, Table 1).

Amphiboles much richer in alumina were found as minor constituents in a corundum and spinel-bearing marble, specimens of which were kindly provided to us by Privatdozent Dr. H. Bank of Gebrüder Bank (Gem Cutting and Trade, Import and Export of Gems and Minerals, at Idar-Oberstein, Germany). The small rock samples were labeled "Pakistan," but the exact locality of the material is unknown. According to the microprobe data presented in this paper, these amphiboles are intermediate between the pure pargasite and tschermakite members; they are distinguished on a half-unit cell basis by low Si (about 5.8), high Aliv (about 2.2), high but variable Alvi (ranging from 1.12 to 1.47), low to extremely low Fe (from 0.07 to 0.006), and the absence of minor elements except for Ti.

# **Description of Specimens**

The coarse-grained marble that contains the amphiboles is white to light gray or even dark gray and composed mainly of xenoblastic calcite grains. Red corundum and pink, violet, or blue spinel are the most outstanding abundant accessory minerals. Minor silicate phases include phlogopite and margarite, corundophyllite, Al-rich pargasite, anorthite, sphene, and tourmaline. In addition, rutile and pyrite are also present. Detailed mineralogical work on this interesting paragenesis is in progress and the results will be published elsewhere.

In the amphibole-bearing specimens, the following mineral assemblages were recorded: Specimen AA-5, Calcite—corundum—phlogopite—margarite—corundophyllite—Al-rich pargasite—anorthite—sphene—pyrite; Specimen HA-1/2, Calcite—Corundum—phlogopite—Al-rich pargasite—anorthite—rutile—pyrite.

The amphibole prisms are mostly idioblastic with dominant  $\{110\}$  and are surrounded by calcite, some of them being intergrown with phlogopite and/or margarite. The grain size parallel to the b-axis ranges from about 0.3 to 1 mm in specimen AA-5, and from 1 to 6 mm in specimen HA-1/2. The total

amount of pargasite is estimated at <5 vol percent. Exact data cannot be obtained because of the irregular distribution of the amphibole and the small size of the specimens.

# **Optical and Physical Properties**

Megascopically, color of the amphibole is somewhat different in both specimens. Thus, using the GSA-Rock-Color chart, amphibole AA-5 is moderate yellow green (5 GY 6/4) and HA-1/2 is light olive brown (5 Y 6/6). For specimen HA-1/2, the hardness equals  $5\frac{1}{2}$  and there is good {100} cleavage at  $56^{\circ}$ . In the latter specimen, chips of appropriate thickness (about 0.5 mm) were obtained that show a faint but distinct pleochroism:  $\gamma'$  (pale grayish orange: 10 YR 8/4)  $< \alpha'$  (grayish yellow: 5 Y 8/4). In thin section, however, both amphiboles are nearly colorless. A summary of optical and physical properties is given in Table 1.

The refractive indices of amphibole HA-1/2 were determined using mixtures of 1-mono-bromo-naphthalene and butylcarbitol as immersion liquids and filtered monochromatic light (PAL filter 589 nm). For an exact orientation of  $\alpha$ ,  $\beta$ , and  $\gamma$ , the amphibole fragments were mounted onto a universal stage. The results agree with a linear extrapolation of the best fit curves given by Deer et al (1963, Fig. 77) for tschermakitic amphiboles. In the pargasite-ferro-hastingsite diagram of Deer et al (1963, Fig. 78), the composition-refractive index curves are slightly straightened by our data.

In contrast, the optic axial angle  $2V_{\gamma}$ ), which varies between 50° and 55°, appears to be rather low compared to data of other known tschermakitic and pargasitic amphiboles (Deer *et al.*, 1963, Fig. 77) although similar to values given by Tröger (1971). So far, the lowest  $2V_{\gamma} = 56^{\circ}$  was reported by Serdiuchenko in 1954 (*cit.* Leake, 1968, analysis 538) from

TABLE 1. Optical and Physical Data of Al-Rich Amphiboles

Specimen:	HA-1/2	AA-5	AA-5	AA-5 3	
Grain:		11	2		
α	1.636 ±0.001				
ß	1.639 ±0.001			~-	
1.652 ±0.001					
γ - a (diff.)	0.016 ±0.002				
γ - a (ret.)*		0.017 ±0.001	0.014 ±0.001		
2V (obs)	54° ±2°		51° ±2°	53° ±2°	
2V (obs) 54° ±2° Y (calc) 55° ±1°			50° ±1°		
Dispersion	v > r		v > r	v > r	
Y ^ 0	24° ±2°	23° ±3°	24° ±3°		
OAP	(010)	(010)	(010)	(010)	

a pargasite occurring in a spinel-bearing marble. This amphibole has a lower Mg/Fe ratio (Niggli mg =  $Mg/(Fe^{3+} + Fe + Mn + Mg) = 0.86)$  than the amphiboles HA-1/2 (mg = 0.98,  $2V_{\gamma (obs)}$   $54^{\circ})$  and AA-5 (mg = 1.00,  $2V_{\gamma \text{(obs)}}$  51-53°). Thus a negative correlation between  $2V_{\gamma}$  and the Mg/Fe ratio as indicated by Tröger (1971, Fig 186-2) seems to be stipulated. For both amphiboles, the 2V values directly determined on the universal stage (using conoscopic control) and calculated from retardation measurements (universal stage, Berek compensator, PAL filter) agree very well with each other (Table 1). The same holds true for birefringence as determined by the calculated difference  $\gamma - \alpha$  (HA-1/2) and by retardation measurements. In the latter case, thickness of the section was obtained by measuring retardation of corundum in the same specimen (AA-5). Since none of the amphibole grains are twinned, the extinction angle  $\gamma/c$  had to be determined by measuring one or a pair of cleavage planes on the universal stage, a method which is not very accurate although the results compare well with other tschermakitic and pargasitic hornblendes (Deer et al, 1963, Table 42).

### X-Ray Data

X-ray film data were obtained from amphibole HA-1/2 by a 114.59 mm camera with Ni-filtered Cu radiation. The sample was identified as amphibole by comparison of the d-spacings with those listed for pargasite and hornblende on JCPDS-PDF cards #11-493 and #9-434, respectively. The d-spacings were initially indexed by analogy to the hornblende data since pargasite d-spacings on JCPDS-PDF card #11-493 are not available above 3.35. Several discrepancies in the comparison led to the use of the least-squares unit cell refinement computer program of Evans et al (1963). This was done by keeping the indexed reflections fixed and allowing the program to select indices for the unindexed reflections. A total of 77 reflections were distinguished in the film. Refined X-ray data for amphibole HA-1/2 give cell parameters of a 9.849  $\pm$  0.002, b 17.927  $\pm$ 0.005,  $c 5.308 \pm 0.004$ ,  $\beta 105^{\circ}12' \pm 3'$ ,  $\alpha \sin \beta$ 9.504, and cell volume 904.3  $\pm$  0.6. These values correspond to those given for calcium monoclinic amphiboles (Deer et al, 1963).

## **Chemical Analysis**

Microprobe Analyses. A total of six amphibole grains in sample AA-5 and eight grains in HA-1/2

were analyzed with an ARL-EMX electron microprobe. Mineral standards (three wet-chemically analyzed hornblendes) close in composition to preliminary analyses were used for final analyses. Corrections to the data were made for drift, background, and deadtime; further corrections were made only for Al<sub>2</sub>O<sub>3</sub> content as other elements were very similar in content to the standards. Measurements were made at 15 kV accelerating voltage and 0.02-0.04  $\mu$ A sample current. Analytical accuracy, expressed as weight percentages of the amounts of oxide present are: SiO<sub>2</sub>, TiO<sub>2</sub>, FeO, CaO  $\pm$  0.9 percent; MgO  $\pm$  2.0 percent; Na<sub>2</sub>O and K<sub>2</sub>O  $\pm$  2.5 percent; Al<sub>2</sub>O<sub>3</sub>  $\pm$  3.2 percent and F  $\pm$  10 percent.

Analyses are given in Table 2, together with the structural formula, for the average of amphiboles AA-5 and HA-1/2 from each of the two samples. Sum of the cations, based on (O, OH, F) = 24.00, range between 15.80 and 15.96. These amphiboles may be adequately described by the structural formula  $W_1X_2Y_5Z_8O_{22}(OH, O, F)_2$  where W = Na, K; X = Ca, Y = Mg, Al, Ti, Fe, and Z = Si, Al.

Water Determination. Sufficient material was available in specimen HA-1/2 to separate an amphibole fragment of approximately 150 mg. The sample, after being crushed to a grain size of about 0.5 mm, was purified by handpicking and dissolving the remaining calcite in acetic acid. The combined H<sub>2</sub>O was determined by the method of Lindner and

TABLE 2. Electron Microprobe Analyses (in wt percent) and Structural Formulae of Al-Rich Amphiboles

				Structural Formulae		
	Wt Pe	rcent	(0, OH, F, C1 = 24)			
	HA-1/2	AA-5		HA-1/2	AA-5	
SiO <sub>2</sub>	40.7	41.1	Si	5.75 2.25 } Z=8.00	5.74	
-			Al IV	2.25 } 2=8.00	2.26 \$ 2=8.00	
A1203	20.2	22.6	177			
2 3			A1 <sup>VI</sup>	1.12	1.47	
T10,	2.54	1.53	Ti	1.12 0.27 0.07 3.62	0.16 Y=4.99	
FeO	0.58	0.05	Fe	0.07	0.01	
MgO	17.2	16.1	Mg	3.62	3.35	
CaO	13.1	13.3	Ca	1.99	1.99	
Na <sub>2</sub> 0	2.44	2.00	Na	1.99 0.67 0.17	$0.54 \left\{ x = 2.69 \right\}$	
κ <sub>2</sub> 0	0.95	0.91	K	0.17	0.16	
н <sub>2</sub> о <sup>+</sup> *	1.11	(1.5)	он	1.05	1.40	
F .	1.3	0.9	F	0.58	0.40	
C1	0.04	0.07	C1	0.02	0.07	
Subtotal	100.16	100.06				
0 = F,C1	0.55	0.40				
Total	99.60	99.66				

<sup>\*</sup>  ${\rm H_2O}^+$  content in HA-1/2 done by method of Lindner and Rudert (1969);  ${\rm H_2O}^+$  in AA-5 assumed to be  $\sim$  1.5 wt percent.

Rudert (1969) whereby the sample is thermally decomposed in a furnace and the vapor is transported by a stream of dry carrier gas into an automatic dead-stop titration apparatus (Metrohm). Here, the water is determined by the iodometric method using Karl Fischer's reagent. The standard deviation of this method is  $\pm 20 \mu g$ , that is, in the present case, ±2 percent of the amount present. Whereas the bulk of the water content was liberated at about 800-900°C, the sample had to be heated to 1300°C to completely remove water. We have assumed H2O+ content of sample AA-5 to be higher compared to HA-1/2 in view of the dissimilar amounts of F in both amphiboles and lower Ti content in AA-5 (Table 2). Insufficient amounts of AA-5 material were available to perform a water analysis.

### Discussion

Leake (1968) has listed at least ten criteria that must be fulfilled to sustain a "superior" analysis. Both amphibole analyses (Table 2) are consistent with those criteria (except that a water determination was not possible for amphibole AA-5).

The distinction between amphibole AA-5 and HA-1/2 is small, and their proper classification is a crucial question. In fact, both amphiboles are too high in Al<sup>IV</sup> (viz, high Al<sub>2</sub>O<sub>3</sub> combined with SiO<sub>2</sub>) to consider them as mixed crystals strictly intermediate between pure pargasite and pure tschermakite (Figures 1, 2). Thus, every recalculation of the present amphiboles in terms of end-member molecules must end up either with excess free alumina or some hypothetical amphibole molecule oversaturated in Al. For instance, 54.7 mole percent of amphibole AA-5 and 57.1 mole percent of HA-1/2 could be calculated in terms of a hypothetical Al-pargasite molecule,  $(Na,K)Ca_2Mg_3Al^{VI}_2Al^{IV}_3Si_5O_{22}(OH,F)_2$ , which could be derived either by the substitution tion Si 

NaAl<sup>IV</sup> from tschermakite.

Despite the highly aluminous nature of amphiboles HA-1/2 and AA-5, they still conform to the limiting Al<sup>IV</sup>/Al<sup>VI</sup> ratio of calcic amphiboles statistically evaluated by Leake (1965, 1971) and expressed by the diagonal sloping line in Figure 2. A linear extrapolation leads to the hypothetical Al-pargasite molecule. According to these data, an Al<sup>VI</sup> content >2 is very unlikely and Leake (1965) has even placed considerable doubts as to the existence of nearly pure tschermakite in nature. He has reemphasized this argument recently (1971) on the basis of studies

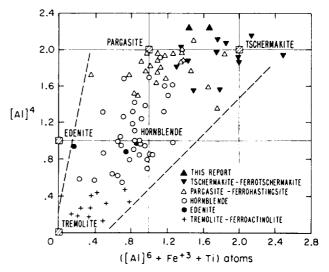


Fig. 1. Plot of Al<sup>1v</sup> vs (Al<sup>vt</sup> + Fe<sup>a</sup> + Ti) in half unit cell of Al-rich Ca-amphiboles compared with different Ca-amphiboles from Deer et al (1963).

on M(2) site occupancy preference by concluding that it is extremely difficult to fill more than 50 percent of M(2) sites with  $Al^{VI}$ . The most Al-rich calcic amphibole known to Leake (1971), and stipulated as being correctly analyzed according to his criteria, contains  $Al^{VI}$  in 67 percent of the M(2) sites; our amphibole AA-5 would have an occupancy of 73 percent Al, if  $Al^{VI}$  were restricted entirely to M(2). This does not appear to be the case for pargasite amphiboles. The calculations of Colville *et al* (1966) yield 17.73 for b if M(2) is occupied principally by

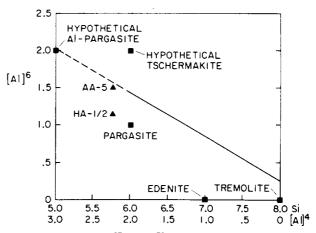


Fig. 2. Plot of Al<sup>1v</sup> vs Al<sup>vi</sup> in half unit cell of Al-rich Ca-amphiboles and theoretical end-members. Diagonal line indicates maximum possible Al<sup>vi</sup> content (Leake, 1965). Linear extrapolation leads to the hypothetical Al-pargasite molecule.

Alvi (synthetic glaucophane II) and 18.05 if occupied entirely by Mg (synthetic tremolite). If a strict correlation between b and the mean radius of the M(2) cations is stipulated and the influence of Na and Ca on the length of the b axis can be neglected (Colville et al, 1966, p. 1741), synthetic Fe-free pargasite, with a theoretical occupancy of Mg1Al1 in M(2) should have b between 17.88 and 17.90. However, synthetic Fe-free pargasite was found to have b = 17.99 by Colville et al (1966), probably indicating partial occupancy of M(2) by  $Al^{vi}$ . Our sample has b = 17.93. If we plot this value on the diagram of Colville et al (1966) that relates length of b axis to radius of the  $M_2$  ion and if we use a linear interpolation between synthetic glaucophane II and synthetic tremolite, we arrive at a mean radius of M(2) ions of 0.60 Å or 40 percent Al and 60 percent Mg. This means that 0.80 Alvi per formula is situated in M(2) while the remainder (0.32) is distributed over the M(1) and M(3) sites of amphibole HA-1/2 (assuming no influence of possible small amounts of Fe or Ti in M(2). In view of the conclusion of Colville et al (1966) who consider a large contraction along b, as produced by the presence of small Al ions in M(2), to be extremely difficult to attain, the definite preference in HA-1/2 of Al<sup>VI</sup> for M(2) rather than M(1) or M(3) sites could indicate a high pressure environment for crystallization. If low pressure synthetic pargasite has less Alvi in M(2) than HA-1/2, then it seems reasonable to assume that  $Al^{vi}$  was stabilized in M(2)through high pressure. This situation may be analogous to experimental studies on glaucophane where the high pressure form contains more  $A^{VI}$  in M(2)than the low pressure form (Ernst, 1963). In addition, the very high amount of Al<sub>2</sub>O<sub>3</sub> in HA-1/2 and especially in AA-5 may be due, in part, to the extremely low Fe contents. Amphibole AA-5 contains the lowest amount of Fe and the highest Niggli value  $[Mg/(Fe^{3+}+Fe+Mn+Mg) = 0.999]$  of any known calcium amphibole. In contrast, sample HA-1/2 has a somewhat lower alumina content and is enriched in Fe by a factor of 10.

#### **Conclusions**

After consideration of the many parameters, we choose to refer to these two amphiboles as Al-rich pargasite or Al-pargasite. The high Al-content of these amphiboles is not too surprising in view of the highly aluminous nature of the environment (corundum—spinel—margarite—anorthite—corundophyl-

lite) in which they crystallized. Total alkali content in both rock samples is probably low; no bulk analyses have been made, but phlogopite is the only major alkali-bearing mineral. Nevertheless, amphiboles still took up sufficient alkali content to preclude formation of pure tschermakite. Leake (1971) concludes from amphibole analyses and theoretical studies that maximum AlVI will occur in amphiboles that crystallized in highly aluminous environments with moderate or low alkali contents and under high pressures, so that amphiboles in corundum-bearing assemblages that crystallized under lower pressure conditions would contain less AlVI than amphiboles in kvanite-garnet-bearing assemblages of higher pressure conditions. In addition, metamorphic amphiboles tend to be more aluminous than igneous amphiboles, which indicates that lower temperatures are also an important factor. This leads to a discussion of aluminous amphibole stability.

Pure pargasite seems to be the amphibole with the highest thermal stability. According to the experimental data of Boyd (1959), the upper stability limit is given by the points 1040°C/1000 bars fluid pressure, 960°C/500 bars, and 850°C/250 bars. At fluid pressures higher than about 800 bars, pargasite melts incongruently to yield Al-diopside + forsterite + spinel + liquid while, at pressures below 880 bars, the breakdown assemblage is Al-diopside + forsterite + nepheline + anorthite + spinel. Tentative experiments by Gilbert (1969) indicate a negative slope of the upper stability curve at high fluid pressures: pargasite is still stable at 20 kilobars/800-900°C, but becomes unstable at fluid pressures close to 30 kilobars giving way to the breakdown assemblages of "sheet silicate" + clinopyroxene + garnet + glass at 800°C and forsterite + clinopyroxene + garnet + glass at 900-950°C. As shown by Boyd (1959), pure pargasite cannot exist in the presence of quartz.

The stability of pure tschermakite is still open to discussion. Boyd (1954) attempted its synthesis although he was not sure that the amphibole composition obtained was stoichiometric. By starting with a glass of tschermakite bulk composition, Gilbert (1969) was able to synthesize an amphibole tentatively identified as tschermakite at 800°C/10 kilobars (uncorrected nominal fluid pressure of the piston cylinder apparatus) with a yield of about 85–90 percent together with minor amounts of garnet. Contrasting results were published by Jasmund and Schäfer (1972) who worked on the tremolite-tschermakite join at fluid pressures of 1, 2,

3, and 10 kilobars. The 10 kilobar runs were carried out by H. S. Yoder in his internal heated hydrothermal apparatus. A gel of tschermakite bulk composition was crystallized at  $800^{\circ}C/10$  kilobars to a mixture of amphibole<sub>88</sub> + chlorite + anorthite. Jasmund and Schäfer (1972) report that, in the entire pressure range of 1 to 10 kilobars, the limiting amphibole composition is tremolite<sub>45</sub> – tschermakite<sub>55</sub>  $\pm$  4 mole percent. Bulk compositions richer in Al than this limit yield amphibole <sub>88</sub> + anorthite + chlorite or amphibole<sub>88</sub> + anorthite + forsterite + enstatite (depending on T and  $P_{11}$ ).

No experimental data are available for the solid solution series pargasite-tschermakite which, of course, would be of special interest for our amphiboles. Moreover, an experimental investigation of the stability of pargasite-tschermakite amphiboles in the presence of a CO<sub>2</sub>–H<sub>2</sub>O fluid phase would be highly desirable.

In conclusion, the present data concur mostly with Leake's contention that highly aluminous amphiboles must form in metamorphic crystallization environments of high aluminum and low alkali contents, and moderate to high pressures; temperatures are probably moderate to high, although a complete study of the paragenesis of more samples is necessary to ascertain if margarite is primary or secondary and if calcite + anorthite + corundum is the major limiting assemblage.

#### Acknowledgments

We thank H. Bank (Idar-Oberstein) for providing the samples, H. Seck (Köln) for stimulating discussions, B. Leake for suggestions and critical comments, V. Rudert and Mrs. U. Meisenburg for advice and help with the water determination, and Mrs. J. Etheridge for assistance in the microprobe data reduction. One of us (T. E. B.) thanks the Mineralogisch-Petrographisches Institut der Universität zu Köln for use of their facilities during a Guest Professor tenure.

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Manuscript received, September 6, 1972; accepted for publication, March 29, 1973.